

Argonne National Laboratory

ORIGIN OF FISSION-PRODUCT RELEASES IN EBR-II,
NOVEMBER 23, 1967, TO MAY 6, 1968

by

R. R. Smith, E. R. Ebersole,
R. M. Fryer, and P. B. Henault

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Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

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EBR-II Project

December 1970

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ABSTRACT

The second verified series of fission-product releases in EBR-II began on November 23, 1967. Results of analyses for ^{133}Xe and ^{135}Xe in the cover-gas system confirmed that the releases originated from fresh fuel material. Attempts to associate the releases with recently installed driver-fuel subassemblies were unsuccessful in view of a subsequent release on December 6, 1967. Eventually the releases were attributed to capsule BC02 in irradiation subassembly X028; capsule BC02 contained a prototypal fuel element of U-Pu-Zr ternary alloy.

The results of postirradiation examinations on capsule BC02 revealed substantial fuel melting and rearrangement, particularly in the upper and lower regions. From these results and from observations of the various fission-product releases from capsule BC02, a physical model of the defect was generated; this postulated model was consistent with all available experimental information.

I. INTRODUCTION

From 1967 to 1969 several inadvertent releases of fission products to the EBR-II primary system have provided important practical information relevant to detection, location, and implications of fuel-element and/or cladding failure in EBR-II. Prior to the first verified fission-product release on May 24, 1967, a great deal of theoretical effort had been devoted to such problems. Although the results of such efforts increased the assurance that the consequences of fuel-element or cladding failure could be tolerated, at least on a short-term basis, substantiating experimental evidence was needed. Such evidence may be acquired in the following three ways: (1) by simulating typical cladding failures in TREAT, (2) by conducting specific experiments under carefully controlled conditions in EBR-II, or (3) by analyzing information provided by inadvertent fission-product releases in EBR-II. Because of the urgent need to acquire a thorough understanding of all aspects of cladding failure in EBR-II and similar fast reactors, all three of these approaches have been used.

Considerable progress has been made by specific experiments conducted in both TREAT and EBR-II, and the results have been reported elsewhere.¹⁻⁶ The present report is primarily concerned with the third approach, the study of inadvertent releases. With the current emphasis on the role of EBR-II as an irradiation test facility, fission-product releases from fuel-bearing materials seem inevitable. In anticipation that other investigators concerned with fuel-failure problems in fast reactors will benefit from a knowledge of similar problems in EBR-II, a policy of documenting all relevant information on EBR-II fission-product releases will be continued.

A. First Series of Verified Fission-product Releases, Beginning May 24, 1967

The first series of verified fission-product releases in EBR-II, beginning on May 24, 1967, was attributed to the failure of an encapsulated, experimental, $\text{UO}_2\text{-PuO}_2$ fuel element (HOV4) located in irradiation sub-assembly X011.⁷⁻⁹ Evidence that indicated failure of the HOV4 capsule included the following: (1) gross physical damage to the fuel and cladding inside the capsule, (2) release of a small but significant amount of radioactive gas when the capsule was placed in an evacuated chamber, (3) a decrease in capsule weight during the irradiation by an amount approximately equal to the weight of the original sodium bond, and (4) an inventory of rare-gas fission products in the capsule less than that expected from calculations based on fuel burnup. Although the exact location of the defect in HOV4 was never physically identified, the fact that all bond sodium had been extruded implied that the defect was very likely in the region of the lower weld.

Of particular interest in this case was the magnitude of the fission-product releases. During the initial release, the signal from the fission gas monitor (FGM) increased by a factor of approximately 3000. Subsequent releases, incurred as the result of operating the reactor in the search mode, were much smaller, i.e., factors of 16, 300, and 220. All releases, however, were characterized by sharp, nearly stepwise increases in the FGM response, followed by simple radioactive decay. Similar behavior was noted for activities of ^{133}Xe and ^{135}Xe in the cover gas. Such behavior suggested a sudden release of a bubble of highly radioactive gas that floated to the surface of the primary sodium and diffused throughout the cover-gas system.

B. Second Series of Fission-product Releases, Beginning November 23, 1967

The second series of fission-product releases began on November 23, 1967. In this series, several unusual phenomena were observed for the first time. Although the initial release on November 23 was characteristic of a simple gas release, disproportionate quantities of ^{133}Xe and ^{135}Xe were found. Whereas significant quantities of the shorter-lived ^{135}Xe were found, the longer-lived ^{133}Xe essentially was missing. This phenomenon was

correctly attributed to the release of newly generated fission products from fresh fuel material. Later symptoms of the release included the generation of secondary buildup curves for ^{133}Xe and ^{135}Xe as their respective iodine parents were continuously extruded with the bond sodium. Much later in the series, presumably as the fuel element deteriorated further, marked fractionation effects occurred. On some occasions, both short- and long-lived, rare-gas fission products were observed; on other occasions, only activities with long half-lives were noted. Such behavior was attributed to consecutive releases: first from the fuel element to the capsule, and then from the capsule to the primary system. If the time between releases was long, the secondary release would be deficient in those activities with the shorter half-lives. If, however, the time interval was short, both short- and long-lived gaseous species would be released.

Finally, bona fide melting phenomena in a metallic fuel element were observed for the first time in the November 23 release. Of importance here was the practical assurance that the consequences of fuel melting could be tolerated without damaging the system. Also important was the demonstration that fuel melting leads to a greatly increased release of volatile fission products.

In general, this report consists of a chronological documentation of events as they occurred. To maintain historical accuracy, the rationale applied in diagnosing the nature of the release on the basis of the available symptoms is given, even though the reasoning proved, on some occasions, to be erroneous.

In the diagnosis of the nature of a given fission-product release, the only information the analyst has at his disposal is that provided by the various fission-product-monitoring systems. Each system provides specific clues which, if taken individually, may mean little. The task of the analyst is to effect a consistency between all available information and a credible physical model.

To avoid the unnecessary repetition of background information, the reader is referred to other sources. A complete description of the reactor plant is given in the original Hazard Summary Report¹⁰ and its Addendum.¹¹ The various fission-product monitoring systems and how the signals from such systems may be used in diagnosing fission-product releases have been discussed by Smith et al.^{12,13}

II. CHRONOLOGY

For simplicity, the chronology of the many releases may be conveniently divided into three individual series which began on November 13, 1967, December 7, 1967, and March 5, 1968. Although each of these series was eventually attributed to the same source, the associated symptoms were so different that each series will be considered individually.

A. The November 23, 1967, Fission-product Release

After the suspect subassembly X011 was removed on June 27, 1967, the reactor was operated for approximately 1900 MWd without any evidence of an additional fission-product release. During this period, all evidence substantiated the conclusion that subassembly X011 was responsible for the releases that occurred during the period May 24 to June 27, 1967.

The next series of releases, from a source other than X011, began on November 23, 1967, during the routine startup of run 26C. Preceding the startup was a series of fuel-handling operations beginning at 0800, November 22. These fuel-handling operations are summarized in Table I.

TABLE I. Fuel-handling Operations
on November 22, 1967

Reactor Grid Position	Subassembly	
	Removed	Inserted
4A2	X015	C2064
4D2	C2000	X015
4B3	C2064	X027
4D3	X016	X028
4F3	C2001	X016
1A1	C283	C2111
4A3	C2002	C2113
6F1	B360	X032
6C1	B3000	X031

Of the various subassemblies transferred at this time, three (X015, X016, and C2064) were in the core prior to the fuel-handling operations. These three were relocated to achieve a more uniform radial temperature distribution across the oscillator rod located in grid position 5A3. Experimental subassemblies X027, X028, X031, and X032 all contained unirradiated, encapsulated, prototypal fuel elements. Driver-fuel subassemblies C2111 and C2113, both containing unirradiated, high-silicon-content fuel, were inserted as a commitment under the fuel-swelling surveillance program.

After the completion of fuel-handling operations, the experimental program called for a measurement of the power coefficient in 5-MWt increments over the 0-45-MWt range. After period calibration of control rods 5 and 12, the reactor achieved criticality at 0513 on November 23, and a power level of 10 MWt was reached at 0609. Power was increased to 15 MWt at 0730 and to 20 MWt at 0839. The FGM alarm point was set at 40% of full scale at 0845.

During the interval 0930 to 0933, two events occurred. First, the power increase to 25 MWt was begun; second, it was noted that the FGM signal was increasing at an abnormal rate. The shift supervisor immediately requested a gas sample (taken at 0940) and noted that no abnormal change was discernible in the FERD signals.* At 0951 the FGM signal continued to increase rapidly and the shift supervisor ordered a power cutback to 17.5 MWt and the evacuation of all personnel from the reactor building. At 1004, the FGM alarmed and a rapid shutdown to 50 kWt was ordered. At 1025, the reactor was brought subcritical. At 1030, the results of gas analyses confirmed that a fission-product gas release had occurred. Restricted entry, under shift-supervisory authorization, was permitted at 1050, since there was no indication of any fission-product release to the reactor building. The FGM was tested with a ^{137}Cs source at 1155 and was shown to be operating satisfactorily. A reconstruction of the strip-chart record for the FGM system during this period is given in Fig. 1. With an assumed time lag of 5-10 min in the response of the FGM, the time of release estimated from the FGM recorder was 0855.

At no time before, during, or after the release was there any indication of an increased FERD signal, reactivity change, unexplained power change, or any off-normal indication in the reading of subassembly-outlet temperatures. (A comparison of critical-position measurements made at 50 kWt before the startup on November 23 and the following day indicated a gain of 1.9 lh, an amount approximately the same as the reproducibility of such measurements.)

From the results of analyses conducted for ^{133}Xe and ^{135}Xe activities in the cover-gas samples taken at 0200 and 0940, it was not immediately obvious that a release had occurred. Significant quantities of these species were present in the cover-gas system, even before the reactor startup. Their presence in the cover gas was the consequence of previous fissions in an ever-present unavoidable contamination of fuel-element surfaces with fuel material. Whereas the FGM system, which senses the presence of relatively short-lived species, registered a large increase, the period of power operation prior to the release was too short to generate large quantities of the longer-lived rare-gas species, i.e., ^{133}Xe and ^{135}Xe .

*FERD, fuel element rupture detector, a device for detecting delayed-neutron-emitting species in a bypass stream of primary coolant.

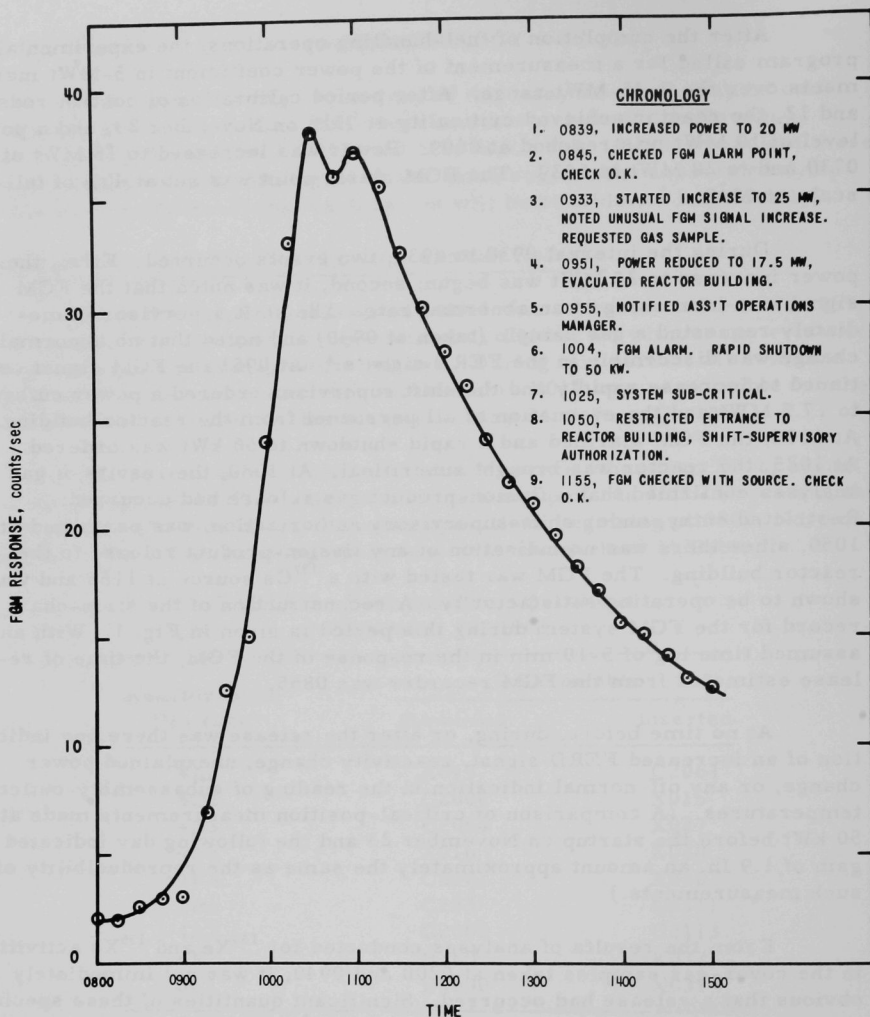


Fig. 1. FGM Response during Original Release--November 23, 1967. ANL Neg. No. ID-103-J5481 Rev. 1.

The preliminary results of an analysis for ^{137}Cs in a sodium sample, collected at 1615 on November 23, failed to indicate a perceptible increase in the ^{137}Cs content of the primary coolant. However, the unusually large amount of ^{137}Cs released to the coolant on May 24 from the mixed-oxide element (HOV4) had greatly increased the normal background level and seriously compromised the usefulness of ^{137}Cs as a fission-product index species in the primary coolant.

proportional to reactor power and to the degree of an unavoidable contamination of fuel-element surfaces with fuel material. Such responses are commonly referred to as the "tramp" background. Thus, even though all fuel elements may be completely intact, small amounts of fission products are generated continuously whenever the reactor is operated. Rare-gas fission products, generated mainly through the decay of their halogen parents, diffuse upward and accumulate in the reactor cover-gas system. Simultaneously, the same rare-gas fission products accumulate in the gas plenum of a given fuel element. The amount, in this case, is proportional to reactor power and to the surface area of the fuel material. If the amount of a given rare-gas index species in the gas plenum of a fuel element is defined as N_{el} , and in the primary cover-gas system as N_{cg} , it follows that the ratio N_{el}/N_{cg} is a constant, regardless of the time-power history of operation. During reactor startup, both N_{el} and N_{cg} increase at identical rates. Furthermore, the ratio N_{el}/N_{cg} should be the same for all gaseous index species.

Based on these considerations, it was tentatively concluded that the release must have originated from fuel material that had been inserted during the shutdown after run 26B. In the belief that additional information could be gained if another release could be initiated, the decision was made to restart the reactor and to bring it cautiously to power.

At 0150 on November 24, the FGM and FERD systems were checked with appropriate sources; both were found to be working satisfactorily. The reactor was restarted, and criticality was achieved at 0657. Power was increased in 5-MWt increments, and a level of 25 MWt was reached at 1416. (After each new power level was reached, a hold of at least 1 hr was imposed.) At 1425, it was noted that the signal from the FGM was again increasing at an abnormal rate. The FGM signal increased from a background level of about 7% of full scale to approximately 30% over a period of less than 2 hr. Coincidentally, a special gas sample was taken at 1422. By 1445, it was concluded that a fission-product release had occurred. At no time preceding, during, or following the release were anomalous changes noted in the FERD response, the power, the reactivity, or subassembly-outlet temperatures. Operation of the reactor continued after the release for approximately 8 hr under 25-MWt conditions. At 2217, the reactor was shut down for fuel-handling operations. A reconstruction of the FGM record during this period is given in Fig. 3, and the activity levels of ^{133}Xe and ^{135}Xe during this period are included in Fig. 2. From the response of the FGM and the results of cover-gas analyses, the time of the release was estimated to be 1420.

An inspection of the data summarized in Fig. 2 revealed that between 1400 and 1422 a relatively large increase occurred in the ^{135}Xe concentration. No perceptible change, however, was noted for ^{133}Xe . Again, this information was considered conclusive evidence that the release must have originated in fresh fuel material inserted after run 26B. Accordingly,

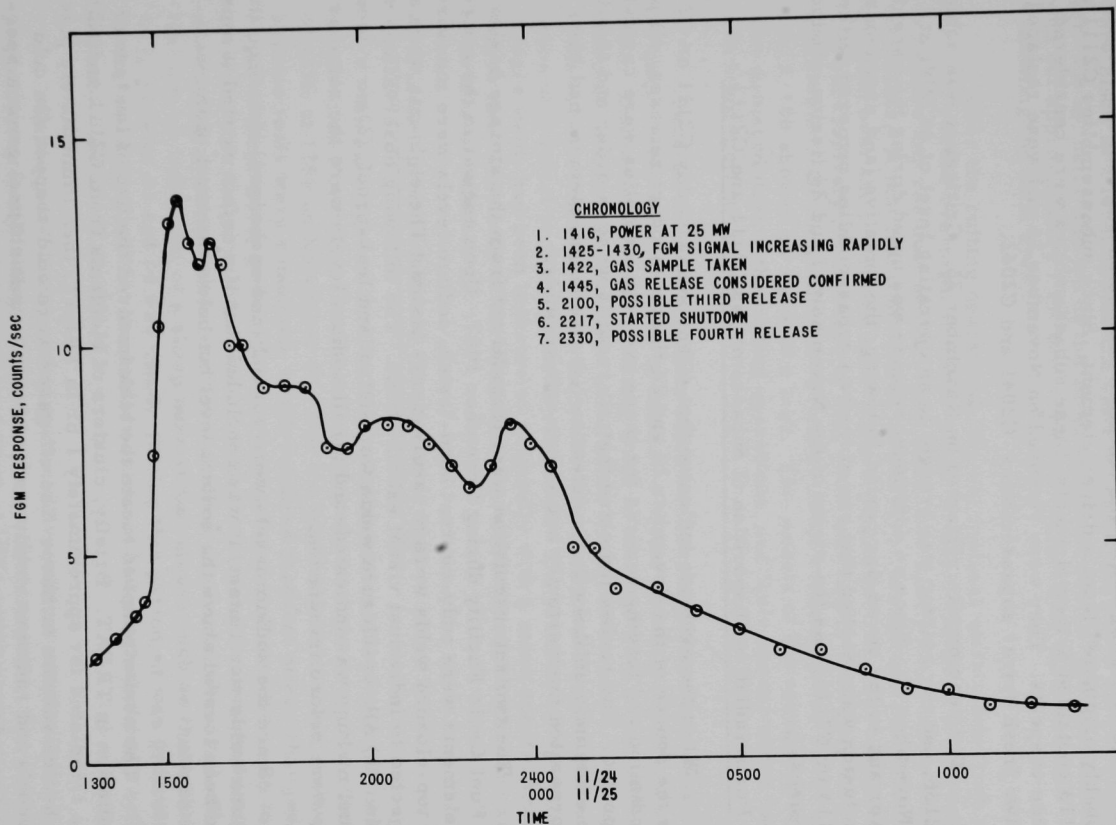


Fig. 3. FGM Response during Second Release--November 24, 1967. ANL Neg. No. ID-103-J5482 Rev. 1.

attention was directed to the subassemblies inserted after the run 26B shut-down (see Table I). Since the release was relatively small (a signal-to-noise ratio of 3.1 for ^{135}Xe), the tentative conclusion was that the release probably originated from a driver element. Since subassemblies C2111 and C2113 contained driver elements, these subassemblies were considered prime suspects. They were removed on November 25 and were replaced by two fresh driver subassemblies, C2041 and C2042.

The reactor was restarted on November 25. Criticality was achieved at 2136, and the reactor was brought to an operating level of 45 MWt at 1359 on November 26. Since no additional release was noted during the ascent to power and in the seven-day period following, the tentative (and erroneous) conclusion was reached that one of the two subassemblies removed (either C2111 or C2113) was the origin of the November 23 and 24 fission-product releases.

B. Postirradiation Disposition of Subassemblies C2111 and C2113

When the evidence indicated that either subassembly C2111 or C2113 was the source of the November 23 and 24 fission-product releases, all pre-irradiation and loading records for these two subassemblies were carefully inspected. The review indicated that all records were in order and that it was extremely unlikely that a defective (i.e., rejected) element had been incorporated inadvertently into either subassembly.

The two subassemblies were transferred from the storage basket to the Fuel Cycle Facility during December 1967. Upon arrival in the air cell, all elements were eddy-current bond-tested, sodium levels were measured, and top-closure welds were pressure-decay tested. The elements were also subjected to individual visual examinations. The following observations were made: (1) All top-closure welds were intact and leak-proof; (2) no evidence of fuel melting was indicated; and (3) all sodium levels were the same as they were before irradiation.

Since the sodium levels were normal (and unchanged) and since the closure welds were intact, it was concluded that if a defect existed it must have been located above the sodium level but below the level of the weld-leak check.

The next attempt to locate the leak involved the use of low-power irradiation in TREAT. Briefly, clusters of elements from C2111 and C2113 were irradiated for approximately 1 hr in TREAT, then immediately placed in a heated vacuum furnace. The off-gases were cold-trapped (on cold charcoal) and radiometrically analyzed with a multichannel gamma spectrometer. As a result of these studies, it was concluded that the defect, if there was one, could not have been large. The effects of a very small defect, however, could have been masked by fission-product gases generated through fission in a small contamination of fuel material on fuel-element surfaces

After all conceivable nondestructive tests had been exhausted, attention was focused on destructive methods. The next step consisted of separating top portions of the jackets from the remainder. Circumferential cuts were made at a level approximately 1 in. below the top of the fuel column (or ~1.6 in. below the top of the nominal sodium level). The spacer wire was cut approximately 1 in. below the top spacer-wire weld to avoid weakening the weld area.

After the cutting operations, the top sections were taken from the air cell, soaked overnight in warm water, drained, resoaked if necessary to remove any residual sodium, redrained, and finally dried under room conditions. A test manifold, based on a 3/16-in. swagelock connector, was inserted over the lower 1/2 in. of a given jacket section. The manifold was then successively pressurized to 500, 1000, and finally 2000 psig with helium under water. Of the combined 182 elements in subassemblies C2111 and C2113, 180 showed no signs of a leak. The jackets of the remaining two had been damaged during the cutting operations, and their spacer wires had been torn from their upper welds.

Because these two jackets could not be tested with the pressurization technique, they were checked by intensive visual and microscopic examinations. Examination of the resulting holes in the weld area revealed that all the metal around the hole area had freshly fractured surfaces. The condition of the surfaces indicated that the welds very likely were sound, and no evidence of a defect in the weld area was noted. The jackets, except for the closure weld, had been leak-tested originally with a mass spectrometer before final assembly of the elements; therefore both sections were assumed to be free of defects.

C. Fission-product Release, December 6, 1967

As discussed in Section II.A, subassemblies C2111 and C2113 were removed on November 25, and the reactor was brought to a power level of 45 MWt at 1359 on November 26. For the following three days, various physics tests were conducted, which required frequent power changes. Because of these many power changes during the period November 26 to December 1, variations in the signals from the various fission-product monitoring systems were expected and were, of course, observed. Nevertheless, no indication of a sharp burst-type release such as those observed on November 23 and 24 was noted. During this period, it was difficult to avoid the conclusion that the earlier releases had originated from either subassembly C2111 or C2113.

On the other hand, rather inconclusive evidence indicated that fission products were still being released to the primary coolant. For example, the equilibrium (saturated) background level of ^{135}Xe in the cover gas had been established at $8.0 \times 10^{-3} \mu\text{Ci/ml}$ during run 26B (when no leaker was in the core). During that portion of run 26C when ^{135}Xe was again saturated, on

December 3-7, the level had increased to $10.2 \times 10^{-3} \mu\text{Ci/ml}$. Since a total of six subassemblies had been added to the core at the start of run 26C, it was tempting to believe that the indicated increase in the ^{135}Xe concentration was the result of an increase in the contamination level of fuel material on the surfaces of the added subassemblies. If such were true, proportionate changes should have been noted in the outputs of the FGM and FERD systems. Accordingly, a careful search of the FGM and FERD records was made. The results of a search dating back to the start of run 26 revealed no discernible change, thus indicating that no change had occurred in the general contamination level of core components with fuel material.

Because of the inability to reconcile the indicated increase in ^{135}Xe activity with an increase in normal fuel-contamination level, attention was directed to the possibility that the source of the November 23 and 24 releases was still in the core. Presumably the nature of the defect had changed, and instead of rare-gas fission products, bond sodium containing chemically fixed ^{133}I and ^{135}I was being extruded. The ^{133}Xe and ^{135}Xe formed from the decay of these species would then account for the increase noted during run 26C. The fact that the FGM and FERD systems failed to sense an increase was attributed to the inability of short-lived halogen fission-product species to survive the trip from their place of recoil birth, through the sodium bond, and out through the defect. The very short-lived iodine and bromine species are sensed directly by the FERD system; their rare-gas daughters are sensed by the FGM.

Activity levels for ^{133}Xe and ^{135}Xe in cover-gas samples during the period December 1 to 13 are summarized in Fig. 4. An inspection of the data reveals that the ^{135}Xe activity reached an equilibrium level during the period December 3 to 7 and that the longer-lived ^{133}Xe was approaching an equilibrium level. During this period, the reactor was operated at 45 MWt in a continuous manner.

The data in Fig. 4 also reveal that some time late on December 6 both activities began increasing and continued to increase until the final shutdown of run 26 on December 11. During the period December 6 to 11, no perceptible increase was noted in the signals from either the FGM or FERD systems, thus indicating that the fuel-contamination level had remained constant. Accordingly, it was concluded that the increased activity levels of ^{133}Xe and ^{135}Xe were the result of a slow continuous extrusion of bond sodium containing significant quantities of ^{133}I and ^{135}I .

D. Fission-product Release, March 5, 1968

The shutdown on December 11, 1967, marked the termination of run 26. During the period between December 11 and the startup of run 27 on February 5, 1968, several changes were made in the core. For the most

part, the changes involved normal end-of-run, fuel-handling operations, i.e., replacement of spent driver subassemblies by fresh ones. Other changes included removing two experimental subassemblies, XG05 and XA08, from the core. These two were considered the next most likely suspects among the subassemblies loaded after run 26B.

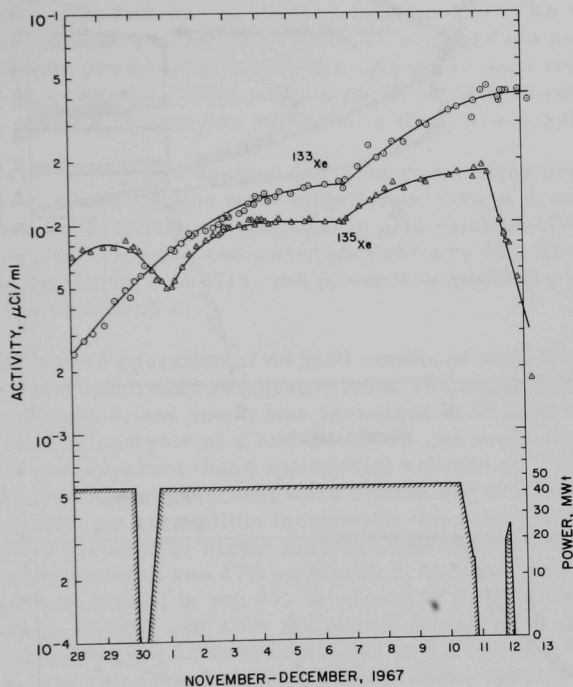


Fig. 4. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--
November 28 to December 13, 1967

Run 27A began on February 5 and terminated on February 29 after 283 MWd of operation. During this period, no evidence of a gas-type release was noted. On the other hand, considerable evidence indicated that bond sodium was still being released through a defect. Activity levels for ^{133}Xe and ^{135}Xe during this period are summarized in Figs. 5 and 6. Of particular interest are the increases noted for both species after the scram of February 9 (see Fig. 5). In this case, the primary pumps were turned off at 0201. Although the reactor was shut down, both activities continued to increase. Presumably, the decrease in primary-coolant pressure permitted the extrusion of bond sodium into the primary system. Since the bond sodium contained both ^{133}I and ^{135}I , their daughters were eventually sensed in the primary cover-gas system.

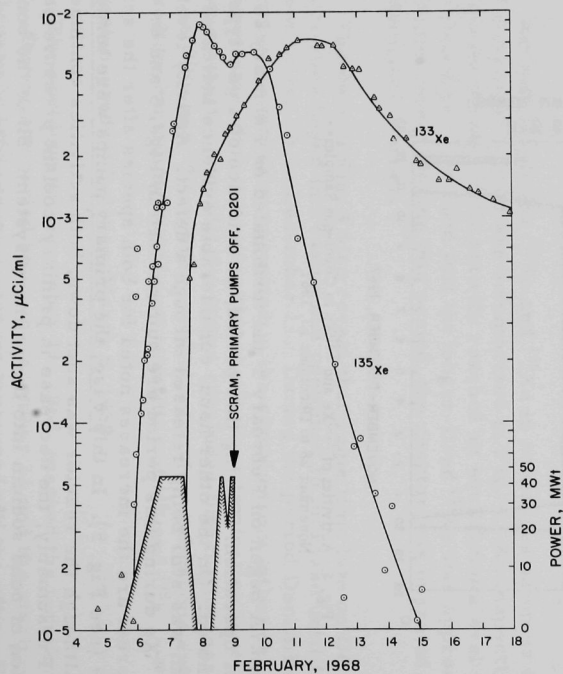


Fig. 5. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--February 4-18, 1968

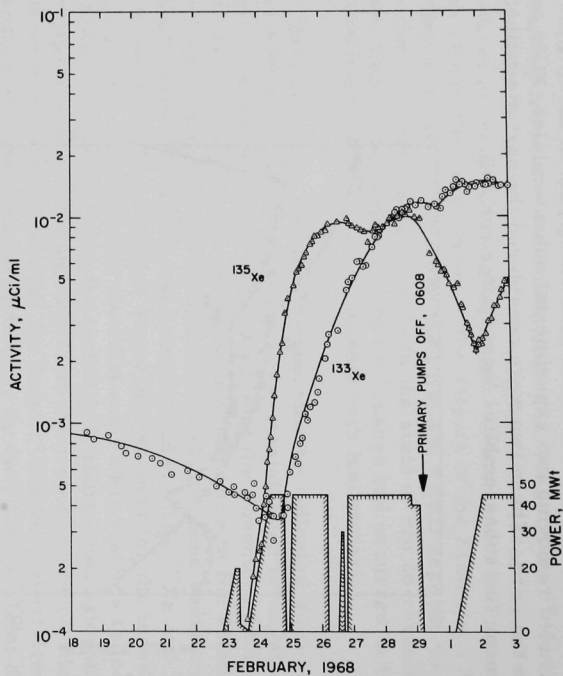


Fig. 6. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--February 18 to March 3, 1968

Another example of this same phenomenon appears in Fig. 6. At 0335 on February 29, the reactor was shut down. Shortly thereafter, at 0608, the primary pumps were secured. Despite this, the ^{133}Xe activity in the cover gas continued to increase for approximately 48 hr. In this case, too, the delayed release was attributed to the extrusion of bond sodium after primary-pump shutdown. The fact that no delayed increase for ^{135}Xe was noted was attributed to a mechanism that preferentially released the parent of ^{133}Xe . Such a mechanism presumably involved a release of bond sodium from a fuel element to a capsule. Then, after a period of days during which the ^{135}I decayed, capsule sodium was released to the primary system.

On February 29, the reactor was shut down, three prototypal re-orificed driver subassemblies were installed in rows 4, 5, and 6, and the suspect subassembly XG05 was installed in grid position 4C2. Fuel-handling operations for run 27B were completed on February 29. The reactor was made critical on March 1 at 0715, and power was gradually increased to 45 MWt at 0030 on March 2.

Steady-power operation at 45 MWt continued until March 5 when, during a series of reduced-flow physics tests, the reactor scrambled. The reactor was restarted, and power was increased to 45 MWt at 1440 on March 5. From an analysis of a routine cover-gas sample taken at 1545 on March 5 it was apparent that a substantial release of ^{133}Xe and ^{135}Xe had occurred. Accordingly, operations were terminated at 2001 on March 5. During this period, no perceptible increase in the output of the FERD was noted. A barely perceptible signal increase was noted, however, for the FGM. At the shutdown of run 27B on March 5, approximately 460 MWd of operation had been logged in run 27. A record of the ^{133}Xe and ^{135}Xe activities before, during, and after the March 5 release is given in Fig. 7. The step increases in the activity levels for both ^{133}Xe and ^{135}Xe were attributed to a gas-type release. The generation of these species through the release of their parents, ^{133}I and ^{135}I (contained in the sodium bond), would have been sensed by gradual buildup curves (for example, the behavior noted during the period December 6 to 11, Fig. 4).

Although perceptible signal increases were not noted for the FERD and FGM systems, the increases for ^{133}Xe and ^{135}Xe in cover-gas analyses were substantial. In the following and all subsequent discussion, release amplitudes will be described in terms of the S/N ratio, i.e., the ratio of the actual (measured) signal strength to the normal background component. In the use of this criterion, care must be taken in evaluating the noise component since this is, of course, a function of the time-power history. Furthermore, all available evidence indicates that the normal equilibrium levels for ^{133}Xe and ^{135}Xe were changing throughout the period November 23, 1967, to May 6, 1968. In early November 1967, before the initial release occurred, normal equilibrium levels of 9.0×10^{-3} and $7.4 \times 10^{-3} \mu\text{Ci/ml}$ were established for ^{133}Xe and ^{135}Xe . In May 1968, after the leaker had

been removed, the respective equilibrium backgrounds for ^{133}Xe and ^{135}Xe decreased to 5.1×10^{-3} and $5.6 \times 10^{-3} \mu\text{Ci/ml}$. In the application of the S/N criterion of signal amplitude, a simple extrapolation of the equilibrium levels between November 1967 and May 1968 has been used. In all cases, the value used for the background component has been based on the extrapolated equilibrium values and the actual time-power history.

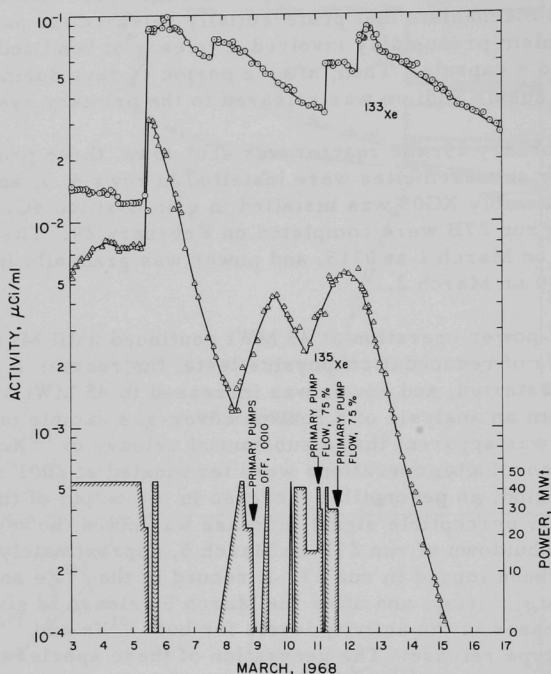


Fig. 7. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--March 3-17, 1968

As an illustration of the use of the S/N criterion, it is constructive to apply this criterion to the ^{133}Xe and ^{135}Xe data given in Fig. 7. The background level for ^{133}Xe immediately prior to the release is indicated to be approximately $1.25 \times 10^{-2} \mu\text{Ci/ml}$, but this value includes a component that originated earlier and had not completely decayed by March 5. The signal strength to be used in evaluating the S/N ratio is nevertheless $8.5 \times 10^{-2} - 1.25 \times 10^{-2} \mu\text{Ci/ml}$, since this is a measure of the March 5 release. The background level to be used is not $1.25 \times 10^{-2} \mu\text{Ci/ml}$, however, since this includes the component from an earlier release. The preferred background value, i.e., from tramp uranium, is $9.0 \times 10^{-3} \mu\text{Ci/ml}$, since ^{133}Xe was essentially at equilibrium at the time of the release. The S/N ratio is then $(8.5 \times 10^{-2} - 1.25 \times 10^{-2}) / 9.0 \times 10^{-3}$ or 8.1.

Using the same considerations, the S/N ratio for ^{135}Xe was

$$\frac{3.1 \times 10^{-2} - 7.4 \times 10^{-3}}{7.4 \times 10^{-3}}$$

or 3.3. The fact that the S/N ratio for ^{133}Xe was larger than that for ^{135}Xe reflects the existence of fractionation phenomena (i.e., a release of gas from a fuel element to the capsule), and then after an indeterminate period of time, a release of gas from the capsule to the coolant. Such a mechanism is also consistent with the fact that only a barely perceptible signal was noted by the FGM system, which senses only very short-lived index species.

Since subassembly XA08 was not in the core during the March 5 release, the evidence suggested its exoneration. Subassembly XG05 was, of course, still suspect. Accordingly, XG05 was removed from the core (position 4C2) and replaced by XA08. The reactor was brought to criticality at 1225 on March 8, but a series of malfunctions in the primary-pump control circuits prevented sustained full-power operation during the period March 8-11.

During reduced-flow experiments at 41.5 MWt on March 11, a small increase was noted in the activity levels for ^{133}Xe and ^{135}Xe . A reactor shutdown was begun at 1515 and completed at 1900. Activity levels for ^{133}Xe and ^{135}Xe during this period are included in Fig. 7.

As with the March 5 release, no perceptible signal increase was indicated by the FERD system, and again only a barely perceptible increase (approximately 30%) was noted in the FGM response. Whereas the March 5 release was relatively strong, giving S/N ratios of 8.1 and 3.3 for ^{133}Xe and ^{135}Xe , respectively, the March 11 release was rather weak. In the latter case, S/N ratios of 7.7 and 0.90 were measured.

Although the reactor was in the shutdown condition, two very small additional releases occurred on the following day. Figure 7 includes the ^{133}Xe and ^{135}Xe activities during this period. Apparently one release occurred between 0500 and 1000, and another between 1430 and 1500. Neither release was sensed by the FGM. Considered particularly significant was the fact that the primary-pump flow was reduced to 75% at 0930 and to zero for the period 1300 to 1451. Apparently, ^{133}Xe gas along with bond sodium was released as the result of the pump shutdown. Approximately one day later another release, involving both ^{133}Xe and bond sodium, took place.

Since XA08 was out of the core during the March 5 release and XG05 was out during the March 11 release, it was tentatively concluded that neither of these was releasing fission products. Attempts to locate the origin of the release after XA08 and XG05 were exonerated were guided by the following knowledge:

1. The decay of ^{135}Xe after the March 5 release followed an approximate 9- to 10-hr decay curve. If bond sodium containing ^{135}I had been released, the decay curve would have been typical of a mother-daughter buildup-decay relationship.

2. The increases in ^{133}Xe and ^{135}Xe counting rates in all cases (March 5, March 11, and two on March 12) were too sharp to identify with a buildup from ^{133}I and ^{135}I activities in the primary sodium system. Step changes in the gaseous activities must, of necessity, originate from a simple gas release.

3. Sodium samples were taken before and after the release on March 5. In this instance, the ^{131}I activity was explainable in terms of a normal buildup from tramp uranium sources. A significant release of bond sodium would have caused a step change in the ^{131}I level of the primary coolant.

4. The failure of the FGM to register a significant increase suggested the preferential holdup of short-lived rare-gas fission products. Such a phenomenon suggested an initial release from an encapsulated fuel element to the surrounding capsule and, after an indeterminate holdup time, a release from the capsule to the primary coolant.

Despite the inability to associate the releases with either XA08 or XG05, the releases were still believed to have originated from an encapsulated fuel element. A release from a driver element could not be reconciled with the relatively strong signal increases.

At the end of run 27C on March 11, approximately 520 MWd of operation had been logged in the run. During the period March 11 to 30, extensive repairs were made to various reactor-control systems; no power operation was logged. During this time, XA08 was removed. Thirteen driver sub-assemblies were also removed and replaced. Figure 8 shows the ^{133}Xe activity during this period.

The reactor was started for the beginning of run 27D on March 30 and was brought to a power level of 45 MWt on March 31. Operations continued normally until 0400 on April 6, when the FGM signal began increasing rapidly. At 0355, the FGM alarmed. Power was reduced to 50 kW at 0356, and the reactor was shut down at 0435.

A reconstruction of the FGM strip-chart record during this period is given in Fig. 9. The data for ^{133}Xe and ^{135}Xe are given in Fig. 10. The S/N ratio for the FGM was 18, and the S/N ratios for ^{133}Xe and ^{135}Xe were 21 and 16, respectively. Following a pump shutdown at 1015, an additional release was observed. In this case, the S/N ratios for ^{133}Xe and ^{135}Xe were 33 and 29. This series of releases marked for the first time a significant increase in the FGM response. As for all previous releases, no increases in the FERD response was noted.

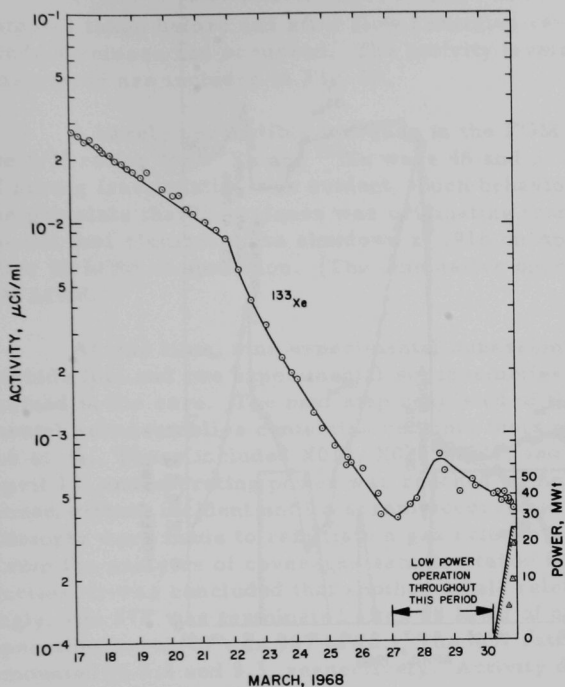


Fig. 8
Activities of ^{133}Xe in Cover-gas
Samples--March 17-31, 1968

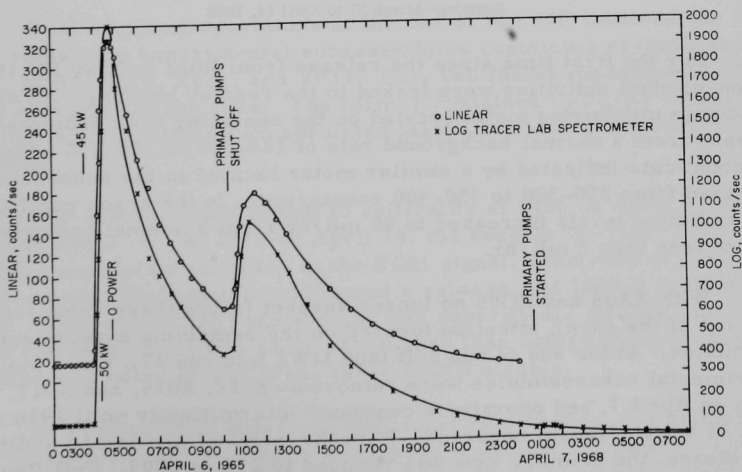


Fig. 9. Reconstructed FGM Record with Subassembly XA08 and Thirteen Driver Sub-assemblies Removed--April 6-7, 1968. ANL Neg. No. ID-103-J5937 Rev. 1.

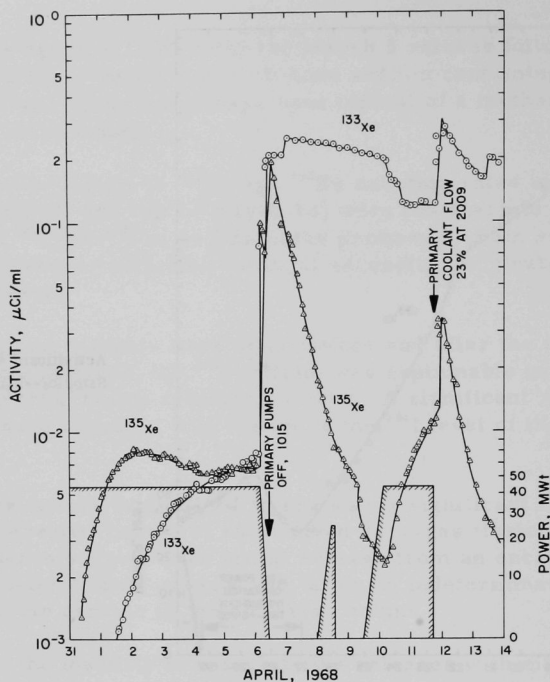


Fig. 10. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--March 31 to April 14, 1968

For the first time since the release from X011 on May 17, 1969,⁹ fission-product activities were leaked to the reactor building. An air-particulate monitoring meter located on the operating floor indicated an increase from a normal background rate of 125-200 to 150-250 counts/min. The count rate indicated by a similar meter located in the subbasement increased from 200-300 to 350-400 counts/min. In the argon purification cell, radiation levels increased to 90 mR/hr from a normal background level of less than 5 mR/hr.

With XA08 and XG05 no longer suspect (since these subassemblies were out of the core), attention focused on the remaining experimental subassemblies. At the end of run 27D (809 MWd into run 27), three more experimental subassemblies were removed--X012, X015, and X017. Run 27E began on April 7, and operations continued intermittently until 1916 on April 11 when a scram occurred. With the intent of causing an additional gas release, the primary flow was reduced to 23% at 2009. Full flow was restored at 2022. A comparison of ^{133}Xe and ^{135}Xe activities in cover-gas

samples taken before and after flow reduction revealed that a small fission-product release had occurred. The activity levels for ^{133}Xe and ^{135}Xe during this period are included in Fig. 10.

A barely perceptible increase in the FGM signal was noted. Since the S/N ratios for ^{133}Xe and ^{135}Xe were 48 and 5, respectively, the existence of strong fractionation was evident. Such behavior tended to substantiate the postulate that the release was originating from an encapsulated experimental fuel element. The shutdown at 1916 on April 11 concluded run 27E after 90 MWd of operation. (The cumulative operation in run 27 was 899 MWd.)

At this time, nine experimental subassemblies containing oxide or carbide fuel and two experimental subassemblies containing metal fuel remained in the core. The next step consisted of the removal of all experimental subassemblies containing ceramic fuels with burnups greater than 0.6 at. %. These included X019, X020, X027, and X032. Run 27F started on April 13, and operating power was reached on April 14. Operations continued without incident until a scram occurred at 0142 on April 16. Again, attempts were made to reinitiate a gas release by reducing the coolant flow. From the analysis of cover-gas samples taken before and after flow reduction, it was concluded that another small release had occurred. Accordingly, run 27F was terminated after 68 MWd of operation. (The cumulative operation in run 27 was 967 MWd.) The S/N ratios for ^{133}Xe and ^{135}Xe amounted to 5.4 and 3.3, respectively. Activity data for ^{133}Xe and ^{135}Xe are summarized in Fig. 11.

Fuel-handling operations at the end of run 27F involved the removal of all remaining experimental subassemblies containing ceramic fuel: XG02, XG03, XG04, X010, X031, and X033. Only two fueled experiments (X028 and X029) remained in the core. The former contained U-Pu-Zr ternary-alloy fuel; the latter contained encapsulated Mark-II uranium-fissium fuel elements.

The reactor was brought to criticality at 1946 on April 17 and to power on April 18. At 1215 on April 19, the console operator noticed a slight but persistent increase in the FGM signal. Analyses of cover-gas samples taken immediately confirmed a release. At 1224 a rapid shutdown was initiated and by 1253 the system was subcritical. At this time the FGM signal had leveled off at a value 28% higher than before the increase. No increase was noted in any of the three FERD signals.

In an attempt to induce an additional gas release, the primary pumps were stopped at 1322 to reduce the pressure against the defect. The results of radiometric analyses of a cover-gas sample taken at 1335 indicated no perceptible increase in the concentration of ^{133}Xe and ^{135}Xe .

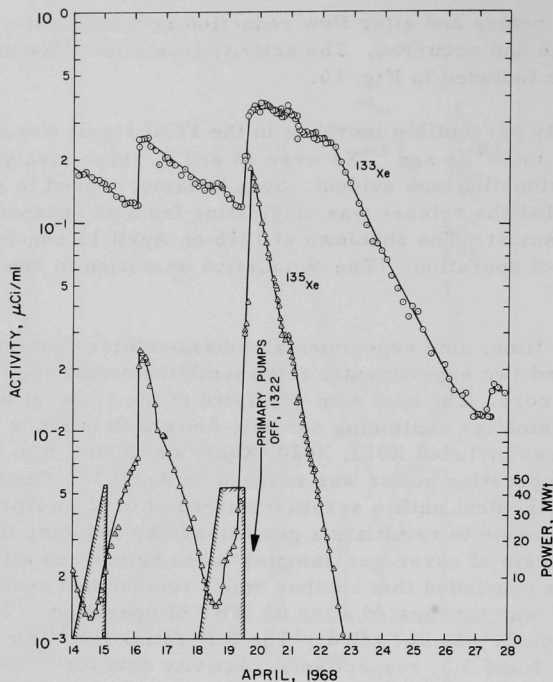


Fig. 11. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--April 14-28, 1968

At 1343 the primary pumps were restarted and brought to full flow. Shortly thereafter the FGM signal began to increase. At 1403 the FGM alarmed and at 1405 the reactor building was evacuated. An air monitor located in the subbasement of the reactor building indicated a tenfold increase in activity during this period. The FGM signal continued to increase, and at 1507 reached a peak value approximately 30 times the normal background level. Figure 12 shows the FGM response during this period. The data for ^{133}Xe and ^{135}Xe are included in Fig. 11. S/N ratios of 88 and 66 were noted for ^{133}Xe and ^{135}Xe , respectively. The shutdown at 1253 on April 19 completed run 27G after 49 MWd of operation. (The cumulative operation in run 27 was 1016 MWd.)

Fuel-handling operations following run 27G consisted of removing the remaining two metallic-fueled experimental subassemblies. The results of analyses conducted for xenon activities before and after the removal of each subassembly revealed an activity increase of 10-20% when X028 was lifted from the core. Such behavior suggested that in lifting X028 (through approximately 10 ft of sodium) the attendant pressure drop caused an additional expansion of gas through the defect.

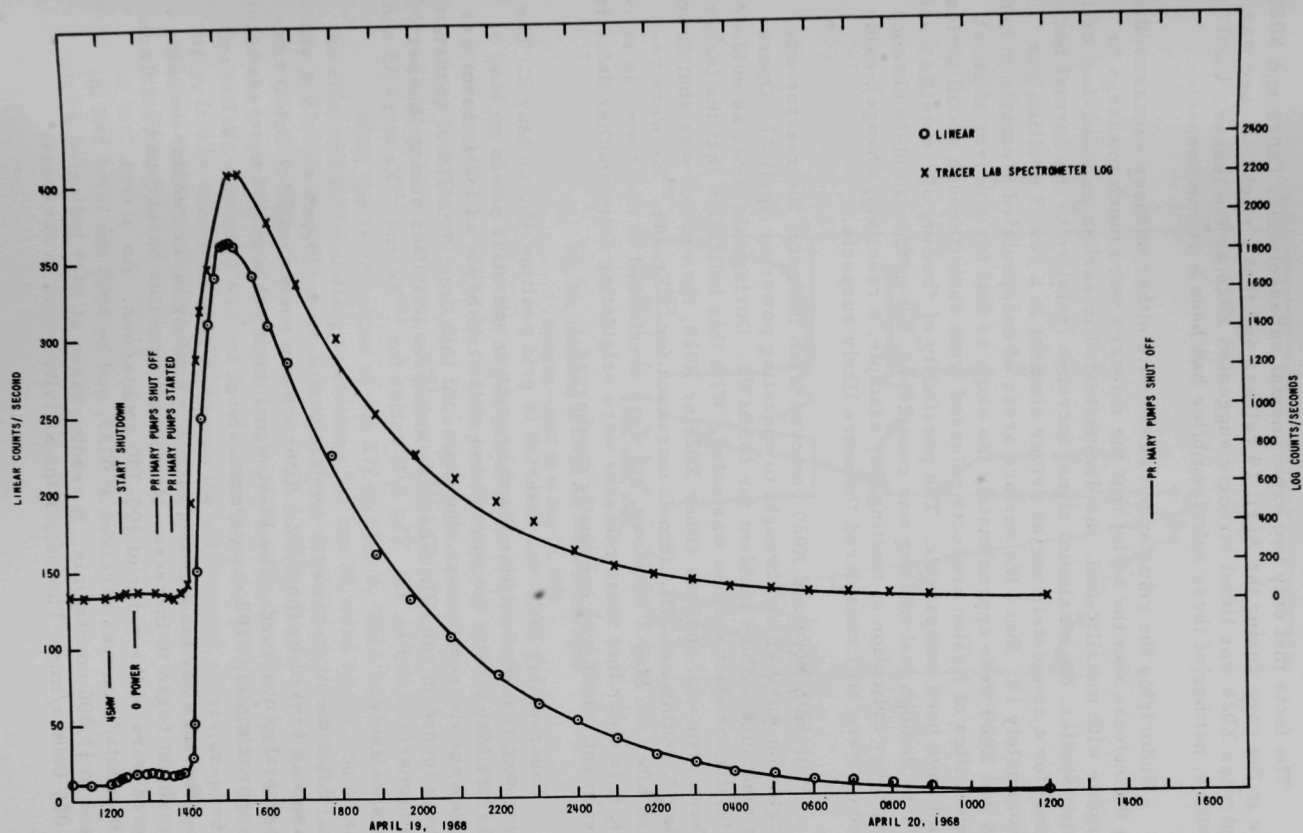


Fig. 12. Reconstructed FGM Record--April 19-20, 1968. ANL Neg. No. ID-103-J5932.

The facts that only two experimental subassemblies (X028 and X029) were in the core during the April 19 release and that a slight release was noted when X028 was lifted strongly suggested X028 as the leaker. Until that time, neither of these subassemblies had been a prime suspect.

Underlying the reluctance to associate either of these subassemblies with the release was the belief that the releases were much too large to reconcile with metallic fuel. As demonstrated in earlier exposed-fuel calibration studies, the maximum signal increase (relative to the normal background) for a completely unclad driver element in a row-5 position was approximately 10. Since the surface areas of encapsulated elements in both X028 and X029 were approximately the same as that for a driver element, the quantities of fission products released from each (through recoil action) should have been comparable. The possibility of "boiling off" volatile fission products through fuel melting was considered, but in the complete absence of any other indication of melting (for example, a reactivity change), oxide elements were still considered the more likely suspects.

With both X028 and X029 removed from the core, the reactor was restarted on April 25 and brought to operating power on April 27. Operations continued without incident for 205 MWd. During this time, no evidence of a fission-product release was noted. With this indication that the leaker had been removed, and was either X028 or X029, the reactor was shut down on May 2, ending run 27H. (Small increases for ^{133}Xe and ^{135}Xe were recorded after the May 2 shutdown, but were overlooked at that time. In retrospect, it is clear that these releases were originating from another defective fuel element, which was eventually discharged.)

Subassembly X028 was inserted in grid position 4D3 on May 2. The reactor was restarted on May 3 and brought to operating power on May 5. At 0539 on May 6, with the power level still at 45 MWt, an FGM alarm was received and the reactor was shut down. At this time, 96 MWd of operation had been logged in run 27I. The ^{133}Xe and ^{135}Xe activities during this period are summarized in Fig. 13. The S/N ratios for ^{133}Xe and ^{135}Xe were 53 and 77, respectively.

After the main primary-coolant pumps were stopped at 0624, a second release was sensed by the FGM. After a pump restart at 0930, a third release was also observed. The FGM signal during this period increased from 15 to approximately 2100 counts/min.

Shortly after the release, airborne activity in the reactor-building subbasement began to increase. Normal air-monitor background levels in this area were of the order of 200-300 counts/min. An alarm, set at 10,000 counts/min, was received at 0755, and by 0900 the level had increased to 13,000 counts/min. A reading taken at 1025 indicated a level of 20,000 counts/min. Thereafter the activity began to decrease.

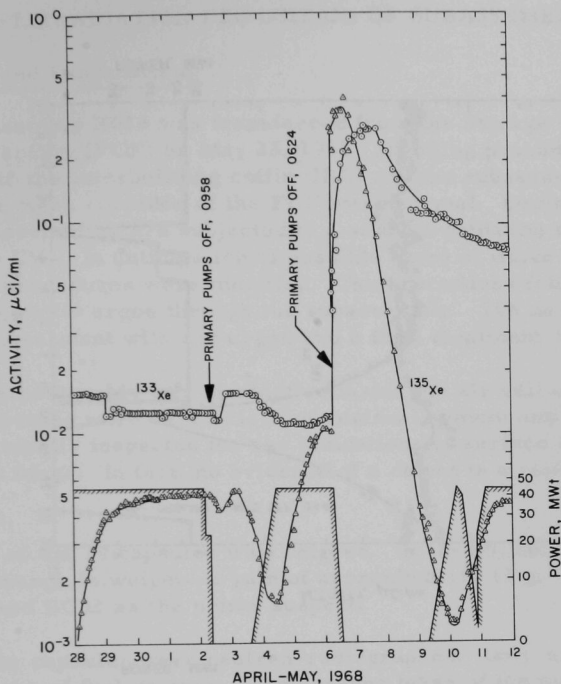


Fig. 13. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--April 28 to May 12, 1968

An alarm from the argon-purification cell, set for 5 mR/hr, was received at 0623. A general survey of the reactor building at 0730 revealed activity levels of 3-5 mR/hr. During the late morning, activity levels decreased rapidly and were essentially unmeasurable by 1800.

With the termination of run 271 on May 6, X028 was removed from the core and fuel-handling operations for run 28 were begun. Subassembly X029 was also reinserted at that time. The reactor was restarted on May 9 and brought to power on May 11. Activity levels for ^{133}Xe and ^{135}Xe for the period May 12 through June 9 are summarized in Figs. 14 and 15. Operations continued without incident during runs 28 through 31. Accordingly, it was concluded that X028 was the sole source of fission-product releases that began on November 23, 1967, and continued intermittently until May 6, 1968.

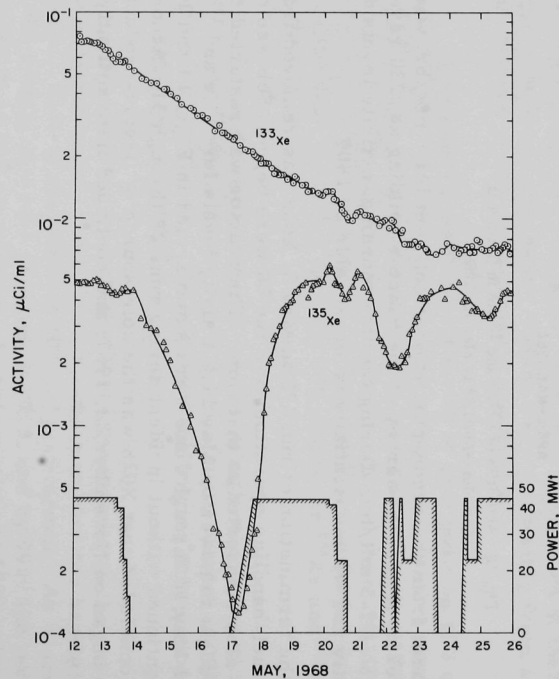


Fig. 14. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--May 12-26, 1968

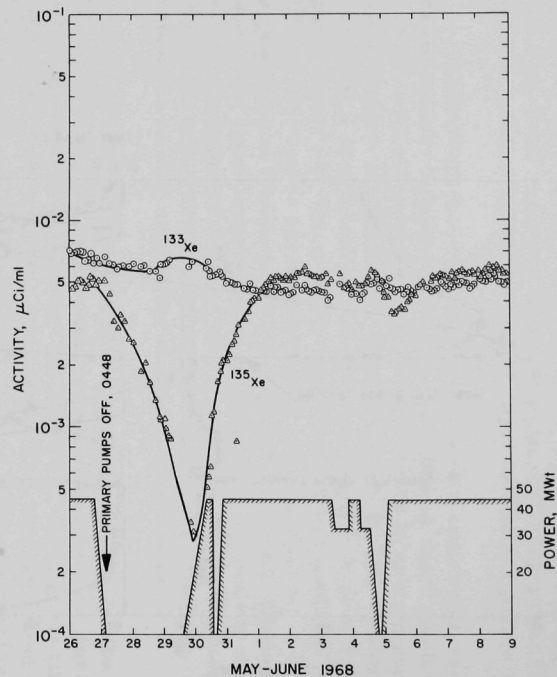


Fig. 15. Activities of ^{133}Xe and ^{135}Xe in Cover-gas Samples--May 26 to June 9, 1968

III. POSTIRRADIATION DISPOSITION OF SUBASSEMBLY X028

A. Handling and Examination

Subassembly X028 was transferred from the storage basket to the Fuel Cycle Facility (FCF) on May 28, 1968. Handling procedures up through the transfer to the interbuilding coffin (IBC) and the subsequent transfer of the IBC to the wash corridor of the FCF were normal. Ordinarily, discharged subassemblies are subjected to a sodium-oxidation water-wash procedure in the IBC. In anticipation of possible entry of water through the defect, wash procedures were modified. The procedures followed consisted of circulating moist argon through the subassembly. The next step consisted of a similar treatment with dry argon and a final treatment with dry air.

The subassembly was dismantled inside the air cell of the FCF. Individual capsules were wiped clean of residual sodium and sodium compounds and visually inspected for any indications of surface damage. No evidence was found. In fact, no evidence of a defect in a capsule was ever found.

Each of the 19 capsules was weighed. Of the 19, only one, BC02, indicated a change in weight--a gain of approximately 11 g. This evidence clearly marked BC02 as the prime suspect.

All the capsules were neutron-radiographed. Only one, BC02, revealed evidence of fuel damage. Radiographs taken of the upper third, mid-section, and lower third are reproduced as Figs. 16-18. The radiographs

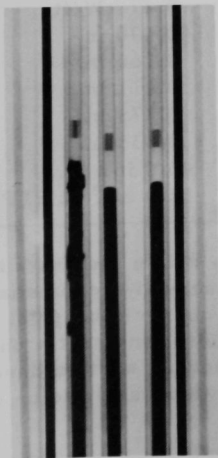


Fig. 16
Neutron Radiograph of Upper
Section of Capsule BC02

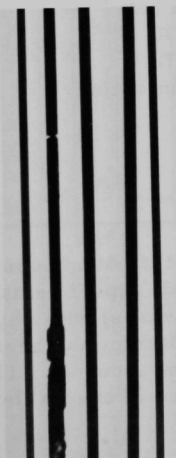


Fig. 17
Neutron Radiograph of Mid-
section of Capsule BC02

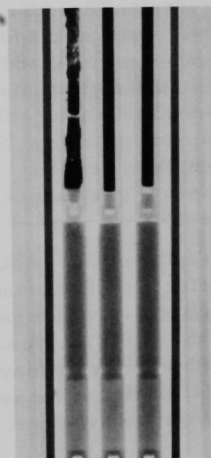


Fig. 18
Neutron Radiograph of Lower
Section of Capsule BC02

show clearly that substantial quantities of fuel at the upper and lower ends of the pin melted and either flowed or slumped outward until the fuel was confined by the colder capsule wall.

B. Description of Capsule BC02

Figure 19 is a schematic diagram of capsule BC02 for reference during the following discussion of a physical model for the fission-product release. The diagram gives the postulated locations of two flaws, one at the upper weld, and the other at the lower.

The fuel material consisted of a ternary metallic alloy having the weight composition 75% uranium, 15% plutonium, and 10% zirconium. Additional details of the fuel element and capsule are given in Table II.

TABLE II. Physical Data for Capsule BC02

Fuel-element length	13.51 in.
Fuel-element diameter	0.144 in.
Weight of fuel material	56.31 g
Volume of fuel material	3.574 cm ³
Density of fuel material	15.75 g/cm ³
Weight of ²³⁸ U	2.91 g
Weight of ²³⁵ U	39.16 g
Weight of plutonium	8.44 g
Fuel-element gas plenum	0.64 cm ³ at STP
Elevation of sodium above fuel column	0.27 in. at STP
Linear power-generation rate	9.47 kW/ft
Fuel-element cladding, OD	0.196 in.
Fuel-element cladding, ID	0.166 in.
Thickness of annular bond sodium	0.011 in.
Capsule cladding, OD	0.375 in.
Capsule cladding, ID	0.335 in.
Length of capsule	40.0 in.
Capsule gas volume	~12.0 cm ³ at STP

Obviously, it is difficult if not impossible to define the exact nature of the defect and to associate all experimentally observed phenomena (principally fission-product gas releases) with physical changes in BC02. Despite these difficulties, an attempt has been made to construct a physical model, which may or may not be accurate. The following section describes a physical model and a sequence of events, both of which seem to be consistent with the evidence.

C. Physical Model of the BC02 Fission-product Releases

Flaw A in Fig. 19 is postulated to have always been present at the lower weld of the fuel element. During hot-bonding op

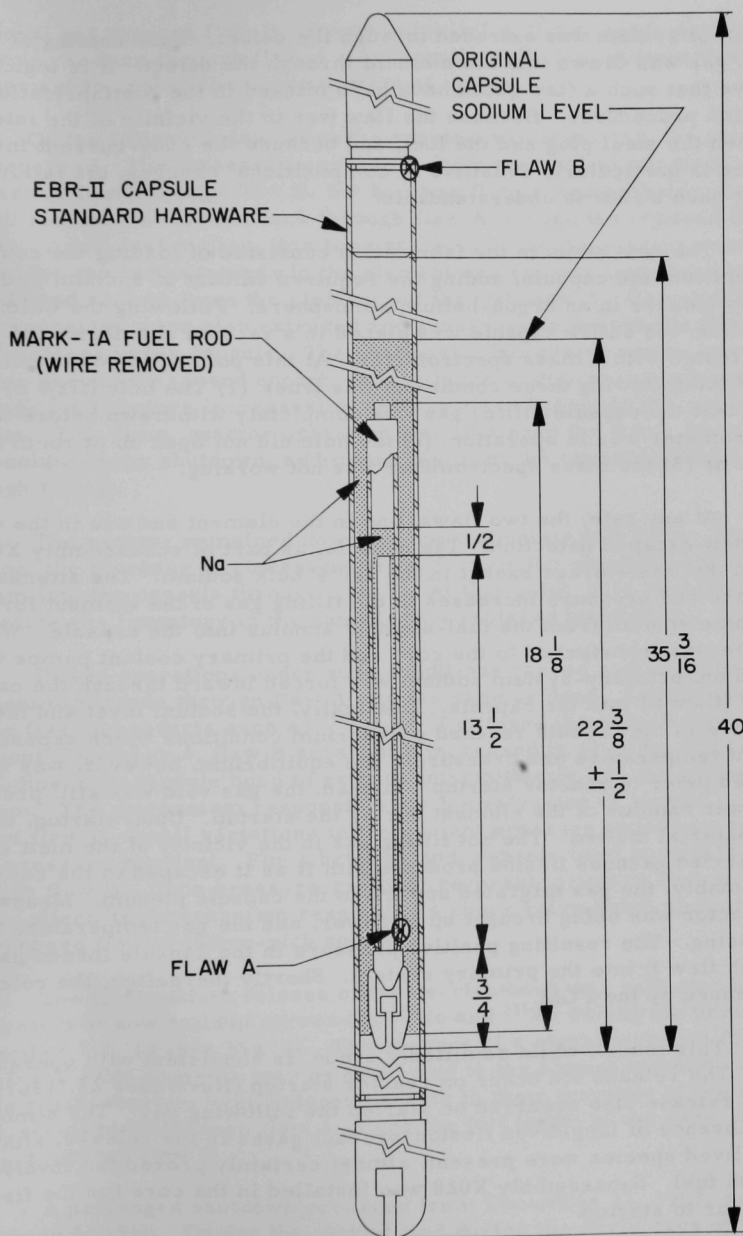


Fig. 19. Diagram of Capsule BC02 Showing Postulated Location of Two Flaws

amount of sodium was extruded through the defect. Upon cooling of the element, gas was drawn into the element through the defect. It is logical to believe that such a flaw could have been missed in the postfabrication inspection procedures. Because the flaw was in the vicinity of the interface between the steel plug and the fuel, and because the eddy-current inspection method is particularly sensitive to compositional changes, the failure to detect such a flaw is understandable.

The next steps in the fabrication consisted of loading the completed element into the capsule, adding the required amount of sodium, and welding the top closure in an argon-helium atmosphere. Following the welding operation, the entire capsule was placed in a vacuum chamber. Off-gases were tested with a mass spectrometer. At this point it is postulated that one of the following three conditions was true: (1) The hole (flaw B) was so large that the capsule-filling gas was completely withdrawn before the mass spectrometer was in operation, (2) the hole did not open up at room temperature, or (3) the mass spectrometer was not working.

At any rate, the two flaws, one in the element and one in the capsule, somehow escaped detection. The capsule, as part of subassembly X028, was loaded into the storage basket in the 700°F bulk sodium. The attendant temperature and pressure increases in the filling gas of the element forced gas and some sodium from the fuel-element annulus into the capsule. When the capsule was transferred to the core and the primary coolant pumps were turned on, primary-system sodium was forced inward through the capsule defect (flaw B) into the capsule. Eventually, the sodium level and the gas pressure in the capsule reached equilibrium conditions which depended on coolant temperature and pressure. The equilibrium, however, was not reached prior to reactor startup. Instead, the gas void was still present in the lower annulus of the element during the startup. Upon startup, unbonded fuel material melted. The hot filling gas in the vicinity of the melt expanded and carried gaseous fission products with it as it escaped to the capsule. Presumably, the gas migrated upward to the capsule plenum. Meanwhile the reactor was being brought up in power, and the gas temperature was increasing. The resulting positive pressure in the capsule forced gas out through flaw B into the primary system. Shortly thereafter, the release was sensed by the FGM.

This model, while admittedly crude, is consistent with operational facts. The release did occur on reactor startup (November 23, 1967). In fact, a release also occurred on startup the following day. The almost complete absence of long-lived fission-product gases in the release, although short-lived species were present, almost certainly proved the involvement of fresh fuel. Subassembly X028 was installed in the core for the first time just prior to startup.

After the first shutdown, the primary pumps were turned off. The resulting positive pressure in the plenums of the element and capsule forced

additional gas through flaw B. Pressure relief in the element was achieved by forcing sodium through flaw A. Apparently sufficient sodium was lost to the element to unbond the upper portion of the pin.

On the following day, when the reactor was restarted, the process was reversed. The increase in pressure from the coolant pumps forced primary sodium through flaw B, but because flaw A was probably small, not enough sodium was forced back through flaw A to bond the exposed fuel at the top. Again fuel melted, this time at the top. As the reactor went to power, the gas temperatures in the element and capsule increased and again forced bond sodium from the element out through flaw A. Chemically fixed halogen species were also extruded and subsequently decayed to their rare-gas daughters in the capsule. The continually increasing power level of the reactor eventually caused enough driving force in the capsule plenum (through temperature increases) to release a gas bubble to the coolant stream. This time, reactor operation was continued for 8 hr. Upon reactor and coolant-pump shutdown, additional gas from the capsule plenum escaped through flaw B.

The reactor remained down for approximately two days. At the next startup, the increase in coolant pressure from pump startup forced primary sodium into the capsule through flaw B. At this point, it is postulated that the entire gas inventory in the capsule was replaced by sodium.

Power operations under such conditions caused sodium in the fuel element to extrude through flaw A into the capsule bond. Longer-lived iodine fission products such as ^{133}I and ^{135}I diffused throughout the bond. On or about December 6, flaw B apparently increased in severity and permitted the iodine-rich capsule bond to extrude intermittently into the primary sodium. The mechanism responsible for the extrusion is simple. At the site of flaw B, small variations in the coolant pressure caused an oscillatory pressure gradient. For a brief period, capsule bond sodium flowed out through flaw B. Upon pressure-gradient reversal, primary sodium flowed in. In effect, the mechanism resembled a check valve which permitted the net outward flow of iodine-rich sodium.

The intermittent release of iodine-rich bond was reflected by the generation of new buildup curves for ^{133}Xe and ^{135}Xe during the period December 7 to 11 (see Fig. 4). The absence of a signal increase for the FERD and FGM systems may be attributed to the almost complete decay of the much-shorter-lived halogen parents in their traverse of the path from recoil birth through flaw A, along the full length of the capsule bond, and out through flaw B.

A prolonged shutdown occurred from December 11, 1967, to February 5, 1968. During this period and during the early days of the run-27 startup, the condition of the fuel element in BC02 deteriorated

considerably. It is likely that additional flaws developed near the top of the fuel pin. This provided a direct gaseous communication between the top of the fuel element and the capsule. Beginning on March 5 and eventually ending on May 6, 1968, a large number of releases occurred. On some occasions, shorter-lived gaseous index species were released; on others, such species were entirely missing. Such behavior seems almost certainly the result of a sequential release: first a release from the element to the capsule, and then, after an indeterminate period, a secondary release from the capsule to the primary coolant system. If the intervening period was short, short-lived gaseous species were seen. If the period was long, only longer-lived species were sensed.

Admittedly, the model described above is conjectural and would be virtually impossible to prove. Nevertheless, it does describe credible physical actions and is at least qualitatively and semiquantitatively consistent with experimental observations.

IV. SUMMARY

The second verified series of fission-product releases in EBR-II began on November 23, 1967. Results of analyses for ^{133}Xe and ^{135}Xe in cover-gas samples confirmed that the releases originated from fresh fuel material. In fuel-handling operations just before the startup, six subassemblies containing fresh fuel had been inserted into the core. Two of these, fuel-surveillance subassemblies, were removed, and the reactor was then operated until December 6, 1967, with no evidence of an additional fission-product release. Exhaustive postirradiation examinations were conducted on the two discharged subassemblies; no evidence of a defect in either of them was found.

Two weeks later, on December 6, 1967, another fission-product release occurred. From this time until May 1968, attention was focused on locating the origin of the release. Groups of experimental subassemblies were removed sequentially, and after each removal, the reactor was operated to initiate an additional release. By a process of elimination, the release was eventually traced to experimental subassembly X028. This subassembly was transferred to the air cell of the FCF and was disassembled for postirradiation examinations. Of the 19 capsules therein, one, BC02 (containing a U-Pu-Zr ternary-alloy element) indicated a weight gain of 11 g. Subsequent neutron radiography of capsule BC02 revealed the existence of gross melting effects, particularly in the upper and lower regions of the element.

From the results of the postirradiation examinations and observations of the behavior of the various releases, a physical model of the release was generated, and this model was shown to be consistent with all experimentally derived information.

ACKNOWLEDGMENTS

We express our sincere thanks and gratitude to the many members of the EBR-II operations group, in particular to F. S. Kirn, J. R. Davis, and G. E. Deegan, for their participation in cover-gas surveillance activities. We also express our thanks to D. C. Hampson, N. R. Grant, and R. V. Strain, all of the Fuel Cycle Facility, for their participation in the postirradiation examination studies.

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